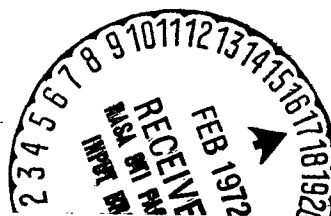


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SILICIC ACID CONTENT IN HYPERBASITES
AND THEIR SERPENTINIZATION

L. V. Dmitriyev and A. V. Garanin

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OCEANIC
SILICIC ACID CONTENT IN HYPERBASITES
AND THEIR SERPENTINIZATION

L. V. Dmitriyev and A. V. Garanin¹

ABSTRACT: For the evaluation of the degree of element removal and addition during serpentinization of hyperbasites, a correlation has been suggested between the ΔSi value, which shows the silicic acid consumption of the rock for serpentine formation and the amount of H_2O . On the basis of 71 analysis of hyperbasites of the Indian Ocean and the Mid-Atlantic Ridges an equation has been obtained derived for the straight line which characterizes ΔSi with H_2O increasing. The comparison of this straight line with lines of possible theoretical ΔSi shows that the serpentinization of oceanic hyperbasites proceeded isochemically.

Investigation of oceanic hyperbasites (hyperbasites of georiftogenic origin [1, 2] was initiated just a few years ago. It has been found that these rocks, including chiefly harzburgites and lherzolites, are petrographically similar to alpine hyperbasites, but have higher silicic acid content and extremely specific geochemical features such as a much higher concentration of certain lithophylic elemental impurities [2, 3].

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The vast majority of the investigated rocks are strongly serpentinized, and since the serpentinization process is completed in an open system with the supply chiefly of water and perhaps other components, the question arises as to whether or not some relationship exists between the degree of serpentinization of these rocks, their high silicic acid concentration and their geochemical features.

The problem of serpentinization and, in particular, problems of removal and addition of silicic acid, magnesium, iron and elemental impurities in this

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*Numbers in the margin indicate pagination in the foreign text.

process, are the center of much attention. Numerous works have been published, both of a review character and in connection with the solution of particular problems. It is not necessary here, therefore, to present an in-depth review of works pertaining to this problem. It should be pointed out, moreover, that no uniformity of opinion exists regarding the question of the removal and addition of elements during serpentization. By way of example we cite the recently published "Trudy II Ural'skogo Petrograficheskogo Soveshchaniya," [Transactions of the Second Ural Petrographic Conference] [4], containing a number of articles from which it follows that removal and addition of silicon, magnesium and iron takes place in some cases, but not in others. The state of knowledge of this subject is very closely scrutinized in an extensive survey article by D. S. Shteynberg et al. [5], included in the above-mentioned collection. It follows from the survey that serpentization is a complex multistage process that there are no reliable general criteria with respect to the degree of serpentization and that the problem of removal and addition of components during serpentization can be solved approximately only for each specific case.

Foreign analysis of serpentization is in about the same state. We cite for example, two recently published works, one of which [6] shows that serpentization is an isochemical process, while the other [7] describes the possibility of the introduction of silicon and alkali and the removal of magnesium/275 and iron.

The foregoing considerations prompted us to attempt to explain how serpentization affected the silicic acid content of the hyperbasites of the Mid-Atlantic and Indian Ocean ridges.

We now have at our disposal 71 analysis of these rocks², performed at the V. I. Vernadskiy Institute of Geochemistry and Analytical Chemistry, Academy of Sciences of the USSR and a few of them taken from the literature. Some of the analysis were published previously [2], and others are summarized in Table 1.

²The analysts are A. V. Lebedkova and M. A. Kremneva

TABLE 1. COMPOSITIONS OF ULTRABASIC ROCKS OF INDIAN OCEAN AND MID-ATLANTIC RIDGES AND THEIR STANDARD MINERALOGICAL CHARACTERISTICS (Niggli).

Component	1		2		3		4		5		6		7	
SiO ₂	38,22	44,49	39,04	45,50	39,00	45,20	38,60	44,40	40,50	45,51	39,54	38,35	39,08	38,16
TiO ₂	0,01	0,01	0,01	0,01	0,05	0,05	0,06	0,07	0,02	0,02	0,07	0,06	0,03	—
Al ₂ O ₃	1,92	2,24	0,73	0,08	1,59	1,84	1,60	1,84	1,71	1,92	1,65	1,88	1,47	1,70
Fe ₂ O ₃	5,48	6,42	6,66	7,72	5,72	6,50	6,43	7,40	4,98	5,59	5,23	3,75	4,95	3,58
FeO	1,80	2,09	1,80	2,10	2,15	2,50	2,50	2,86	3,09	3,47	2,50	1,99	3,00	2,46
MnO	0,09	0,12	0,09	0,10	0,09	0,10	0,11	0,12	0,13	0,14	0,14	0,11	0,11	0,06
MgO	38,10	44,4	38,00	44,40	37,70	43,70	37,50	43,2	38,44	43,19	37,58	52,95	36,84	53,57
CaO	0,20	0,23	—	—	Trace	—	Trace	—	0,05	0,06	—	—	—	—
Na ₂ O	0,08	0,09	0,08	0,09	0,09	0,10	0,08	0,09	0,09	0,10	0,40	0,68	0,20	0,35
K ₂ O	—	—	—	—	0,01	0,01	0,02	0,02	Trace	—	0,18	0,23	0,10	0,12
H ₂ O ⁺	12,90	—	12,00	—	11,50	—	11,30	—	9,10	—	10,30	—	11,50	—
H ₂ O ⁻	1,47	—	1,44	—	1,40	—	1,20	—	1,04	—	1,10	—	1,30	—
Loss on calcination	—	—	—	—	—	—	—	—	—	—	1,53	—	1,11	—
Sum	99,27	—	99,81	—	99,30	—	99,40	—	99,15	—	100,22	—	99,69	—
<i>ol</i>	69,5	—	70,0	—	66,5	—	68,5	—	64,0	—	69,8	—	65,6	—
<i>Hy</i>	25,2	—	27,9	—	29,6	—	27,7	—	31,6	—	20,8	—	28,8	—
<i>Di</i>	3,2	—	1,4	—	1,8	—	1,4	—	1,8	—	8,0	—	3,8	—
<i>Sp</i>	2,1	—	0,7	—	2,1	—	2,4	—	2,6	—	1,4	—	1,8	—

Component	15		16		17		18		19		20		21	
SiO ₂	42,40	46,60	38,90	45,0	39,56	45,1	40,06	45,35	40,26	45,00	40,42	45,00	41,60	47,2
TiO ₂	0,02	0,02	0,11	0,1	0,04	—	0,06	0,06	0,07	0,08	0,09	0,10	0,06	0,1
Al ₂ O ₃	1,56	1,72	2,83	3,3	1,90	2,2	2,40	2,72	2,80	3,14	3,63	4,55	1,33	1,5
Fe ₂ O ₃	4,57	5,02	4,90	5,7	7,30	8,3	6,92	7,81	4,81	5,40	4,59	5,15	7,13	8,1
FeO	3,90	4,30	3,05	3,5	1,43	1,6	2,15	2,44	3,76	4,20	3,05	3,41	2,33	2,6
MnO	0,13	0,14	0,12	0,1	0,73	0,8	0,13	0,15	0,15	0,16	0,17	0,19	0,20	0,2
MgO	37,10	40,70	35,04	40,5	35,32	40,2	34,98	39,64	35,05	39,40	34,70	38,80	33,30	37,9
CaO	1,16	1,28	1,20	1,4	1,20	1,4	1,30	1,50	2,15	2,40	2,20	2,45	0,82	0,9
Na ₂ O	0,20	0,22	0,26	0,3	0,23	0,3	0,25	0,28	0,18	0,20	0,28	0,30	0,93	1,1
K ₂ O	—	—	0,08	0,1	0,11	0,1	0,05	0,05	0,02	0,02	0,05	0,05	0,31	0,4
H ₂ O ⁺	7,20	—	10,90	—	10,20	—	10,40	—	9,00	—	9,20	—	9,60	—
H ₂ O ⁻	0,83	—	1,26	—	1,25	—	1,11	—	1,00	—	1,02	—	1,11	—
Loss on calcination	—	—	0,98	—	1,01	—	—	—	—	—	—	—	1,59	—
Sum	99,07	—	99,63	—	100,28	—	99,81	—	99,25	—	99,40	—	100,31	—
<i>ol</i>	60,0	—	59,7	—	66,0	—	60,6	—	61,1	—	58,0	—	59,0	—
<i>Hy</i>	29,0	—	20,1	—	19,9	—	25,4	—	21,2	—	21,6	—	21,1	—
<i>Di</i>	11,0	—	15,0	—	14,1	—	14,0	—	17,7	—	20,4	—	19,9	—
<i>Sp</i>	—	—	0,2	—	—	—	—	—	—	—	—	—	—	—

Comment: Total composition, in percent by weight, is given in the left-hand column; given in the right-hand column gives the composition converted to anhydrous residue in percent by weight. The analysts were A. V. Lebedkova, M. A. Kremneva, V. I. Vernadskiy Institute of Geochemistry and Analytical Chemistry, of Sciences, USSR). Harzburgites of the Indian Ocean Ridge: 1, specimen 5319/6813; 2, specimen 5319/6814; 3, specimen 5319/6810; 4, specimen 5319/684; 6, specimen DR 6/2; 7, specimen DR 12/1b. Lherzolites of the Indian Ocean Ridge: 5, specimen DR 12/2; 8, specimen 5319/6812; 9, specimen 5324/689; 11, specimen 5324/59; 12, specimen DR 6/1; 13, specimen 5324/685; 14, specimen 5324/687; 17, specimen TR 15/1v; 10, specimen TR 15/1a; 15, specimen DR 30/1v; 16, specimen DR 12/3; 18, specimen 5324/683; 19, specimen 5324/682; 20, specimen 5324/686; 21, specimen DR 30/1. Atlantic Ocean Harzburgites: 22, specimen RG-10/27, specimen RG 7/4; Lherzolites of the Atlantic Ocean: 23, specimen RG-4; 24, specimen RG 7/1; 25, specimen RG 7/2; 26, specimen RG 7/3; 28, specimen RG-2.

Note: Commas indicate decimal points.

TABLE 1. (Concluded)

Components	8		9		10		11		12		13		14	
SiO ₂	39,48	45,40	39,50	45,50	40,00	46,00	38,38	44,12	39,26	45,2	39,18	45,30	38,60	44,40
TiO ₂	0,06	0,07	0,09	0,10	0,02	0,02	0,05	0,06	0,10	0,1	0,06	0,06	0,09	0,14
Al ₂ O ₃	1,61	1,85	1,00	1,14	1,04	1,20	2,33	2,67	1,67	1,9	2,16	2,60	2,50	28,80
Fe ₂ O ₃	5,87	6,73	5,50	6,35	5,08	5,85	6,03	6,92	5,95	6,8	6,68	7,70	5,90	6,80
FeO	2,50	2,87	3,05	3,55	2,50	2,89	2,87	3,29	2,33	2,7	1,79	2,07	2,69	3,12
MnO	0,13	0,15	0,11	0,12	0,14	0,15	0,20	0,22	0,13	0,1	0,41	0,47	0,13	0,15
MgO	37,20	42,8	36,92	42,50	36,50	42,00	36,50	41,95	36,30	41,8	35,42	41,5	35,80	41,30
CaO	—	—	0,38	0,44	1,48	1,74	0,47	0,53	0,58	0,7	0,77	0,09	0,94	1,04
Na ₂ O	0,09	0,10	0,20	0,23	0,14	0,15	0,16	0,18	0,46	0,5	0,16	0,19	0,13	0,15
K ₂ O	0,03	0,03	0,06	0,07	—	—	0,05	0,06	0,20	0,2	0,02	0,02	0,02	0,02
H ₂ O ⁺	11,10	—	11,20	—	11,6	—	11,60	—	11,80	—	11,50	—	10,50	—
H ₂ O ⁻	1,20	—	1,20	—	1,37	—	1,31	—	0,20	—	1,37	—	1,10	—
Loss on calcination	—	—	—	—	—	—	—	—	1,25	—	—	—	0,60	—
Sum	99,27	—	99,21	—	99,87	—	99,95	—	100,23	—	99,52	—	99,00	—
<i>ol</i>	63,5	—	68,5	—	64,10	—	68,6	—	70,2	—	68,2	—	66,1	—
<i>Hy</i>	32,6	—	25,3	—	23,8	—	22,9	—	17,0	—	10,8	—	22,9	—
<i>Di</i>	1,8	—	6,2	—	9,2	—	6,4	—	12,8	—	21,0	—	9,8	—
<i>Sp</i>	2,1	—	—	—	—	—	2,1	—	—	—	—	—	1,2	—

Components	22		23		24		25		26		27		28	
SiO ₂	44,20	37,76	41,46	46,5	40,96	45,8	40,15	45,6	39,24	44,5	38,00	44,6	39,53	45,0
TiO ₂	0,12	0,05	0,08	0,1	0,17	0,2	0,13	0,1	0,21	0,2	0,04	—	0,36	0,4
Al ₂ O ₃	3,50	3,49	4,40	4,9	3,93	4,4	3,77	4,3	5,60	6,2	2,14	2,5	3,24	3,7
Fe ₂ O ₃	1,06	0,67	5,64	6,3	5,03	5,6	5,85	6,6	7,14	8,2	5,86	6,9	6,97	7,9
FeO	6,80	4,87	2,35	2,6	2,50	2,8	2,64	3,0	2,20	2,5	1,03	1,2	1,73	2,0
MnO	0,13	0,10	0,02	—	0,21	0,2	0,15	0,2	0,27	0,3	0,20	0,2	0,22	0,2
MgO	38,80	49,36	31,63	35,5	33,60	37,5	31,97	36,4	31,17	35,3	37,00	43,4	34,25	39,0
CaO	2,40	2,21	3,05	3,4	2,54	2,8	2,54	2,9	1,81	2,0	0,24	0,3	1,32	1,5
Na ₂ O	0,73	1,18	0,63	0,7	0,45	0,5	0,57	0,6	0,53	0,6	0,49	0,6	0,30	0,3
K ₂ O	0,28	0,31	—	—	0,14	0,2	0,22	0,3	0,21	0,2	0,24	0,3	—	—
H ₂ O ⁺	1,50	—	8,90	—	9,10	—	10,50	—	10,40	—	12,80	—	11,20	—
H ₂ O ⁻	0,13	—	1,15	—	1,13	—	1,22	—	1,22	—	1,47	—	1,20	—
Loss on calcination	0,53	—	—	—	—	—	—	—	—	—	—	—	—	—
Sum	100,18	—	99,31	—	99,76	—	99,71	—	100,14	—	99,51	—	100,32	—
<i>ol</i>	67,5	—	50,5	—	56,8	—	57,6	—	54,1	—	73,2	—	60,3	—
<i>Hy</i>	8,9	—	17,9	—	16,4	—	12,7	—	19,3	—	13,4	—	24,1	—
<i>Di</i>	24,5	—	31,6	—	26,8	—	29,7	—	24,6	—	12,4	—	15,0	—
<i>Sp</i>	—	—	—	—	—	—	—	—	2,0	—	1,0	—	0,6	—

Commas indicate decimal points.

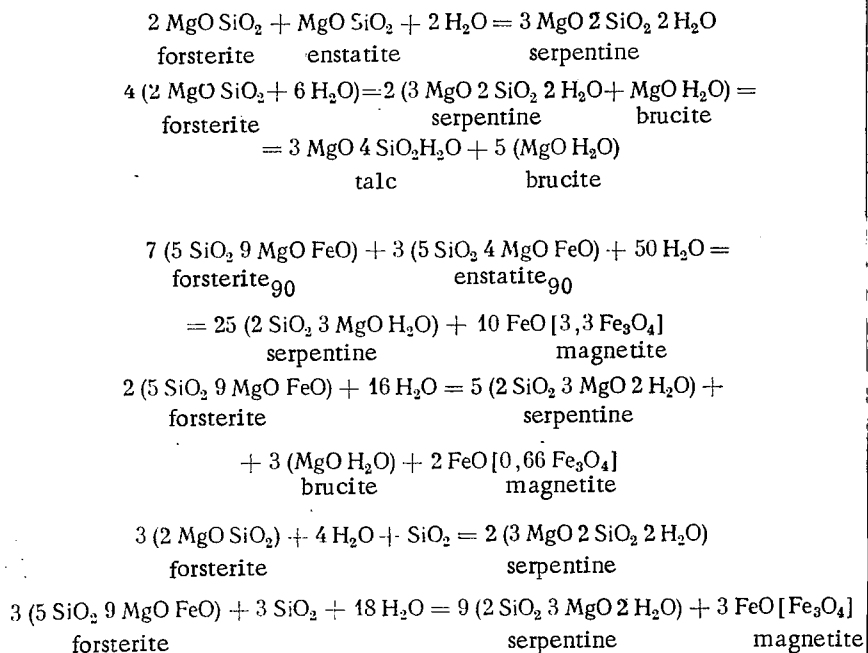
For the solution of the stated problem we proceeded from the fact that as the degree of serpentinization increases the percentage of silicic acid in the primary pyroxenes and olivines of the rocks should decrease by some value proportional to the amount of water bound in the serpentine. This value is

determined according to the serpentine formula: $3\text{MgO} \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ and can be expressed as follows:

$$\Delta\text{Si} = \text{Si} - 0.5\text{H},$$

where ΔSi is the residual atomic silicon bound in the pyroxene and olivine; Si is the total atomic silicon in the serpentinized rock; H is atomic hydrogen, proportional to the percent by weight of H_2O^+ in the serpentinized rock.

The change in ΔSi during serpentinization, proceeding with and without the addition of silicic acid, should be different. To determine the character of this change we used the following possible equations of the serpentinization reaction:



The first four reactions proceed isochemically (with the addition of water only). In reactions (1) and (2) only pure iron-free forsterite and enstatite are serpentinized. Here, as shown by reaction (1), the silicic acid present in the association forsterite-enstatite, taken in the proportion 1:1, goes entirely to pure serpentine without the formation of brucite. At a

higher enstatite concentration the formation of pure serpentine requires the removal of silicic acid or the addition of magnesium. At lower enstatite concentrations, the formation of brucite in association with serpentinite is essential for preservation of the isochemicity of the reaction. | /276

In reactions (3) and (4) forsterite with a concentration of 10% fayalitic component and enstatite with 10% ferrosilitic molecules participate, Conversion of the left-hand sides of equations (3) and (4) to percent by weight yields compositions with FeO concentrations of 10.7% and 8.5%, respectively, i.e., compositions close to the actual hyperbasites with maximum iron content. Here, according to reaction (3), the isochemical character of serpentinization is achieved for a mixture of ferrous olivine and pyroxene, now taken in the proportion 7:3, since the reaction produces magnetite. Reactions (5) and (6) proceed with the addition of silicic acid. | /277

Conversion of the left and right terms of equations (1-6) to percent by weight and to atomic masses and calculation of ΔSi characteristics (Table 2) yield coordinates for the plotting of a nomogram that reflects the theoretical change of ΔSi during serpentinization of ultrabasic rocks with a primary composition from dunites to lherzolites (Figure 1).

The values of ΔSi are plotted on the vertical axis of the nomogram and percent by weight of H_2O^+ on the abscissa. Also shown here is the location of points corresponding to the compositions of nonserpentinized ultrabasic rocks corresponding to the compositions of the left-hand sides of equations (1-6) and the various compositions of completely serpentinized rocks, corresponding to the appropriate right-hand terms in the same equations. The composition of pure serpentine and the composition of serpentine with magnetite lie on the horizontal axis, with the 0 value of ΔSi . The negative values of ΔSi for associations of serpentine with brucite (and with magnetite) are found by virtue of the fact that in the brucite water is bonded with magnesium in the ratio 1:1, but in serpentine water is bonded with the sum of magnesium and silicon in the ratio 2:5. Hence, the water concentration increases in associations with brucite: for pure serpentine $H_2O^+ = 13.0$, but for the association | /278

1 serpentinite + 1 brucite $H_2O^+ = 16.1$. In association with magnetite, the H_2O^+ concentration decreases.

TABLE 2. COORDINATES OF BASIC POINTS ON $\Delta Si - H_2O^+$ DIAGRAM.

Reaction	Association	Oxides	Wt., %	Atomic mass	ΔSi	Number of points on diagram
(1)	Forsterite + enstatite	SiO_2 H_2O^+	47,8 0	796 0	+796	1
(1)	Serpentine	SiO_2 H_2O^+	43,8 13,0	721 1442	0	2
(2)	Forsterite	SiO_2 H_2O^+	43,0 0	716 0	+716	3
(2)	Serpentine + brucite = talc + + 5 brucite	SiO_2 H_2O^+	35,8 16,1	596 1788	-298	4
(3)	7 Forsterite + 3 enstatite / 80	SiO_2 H_2O^+	44,5 0	741 0	+741	5
(3)	25 Serpentine + 3.3 magnetite	SiO_2 H_2O^+	39,3 11,8	655 1310	0	6
(4)	Forsterite	SiO_2 H_2O^+	40,6 0	676 0	+676	7
(4)	5 Serpentine + 3 brucite + 0.66 magnetite	SiO_2 H_2O^+	34,0 16,3	566 1810	-384	8
(2)	Talc	SiO_2 H_2O^+	63,4 4,7	1056 522	+795	9

Note: Commas indicate decimal points.

The straight lines connecting the points of the compositions on the left and right parts of equations (1-4) show the theoretical change of ΔSi during serpentinitization, proceeding isochemically. Associations of serpentinite with talc, enriched with silicic acid, lie above line 1-2 and associations enriched with brucite and impoverished of silicic acid lie below line 7-8. The areas between lines 1-2 and 7-8 should contain the compositions of rocks during whose serpentinitization substantial addition or removal of silicic acid, magnesium or iron does not occur. Isochemical serpentinitization should proceed on any line parallel to lines 1-2 and 7-8 and lying between them. Deviation upward and to the right from this direction obviously must be related to a partial addition of silicic acid (or removal of iron and magnesium), and change in slope

downward and to the right should indicate removal of silicic acid (or addition of iron and magnesium) during serpentinization.

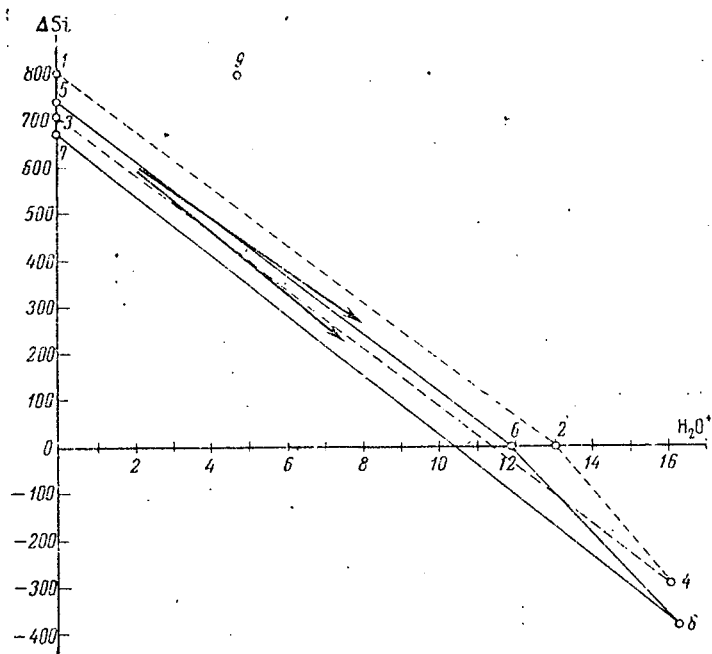


Figure 1. Nomogram of Theoretical Change of ΔSi During Isochemical Serpentinization of Ultrabasites. The numbers of points corresponding to the different mineralogical associations are the same as those in Table 2.

The nomogram can be used in the given form only for a series of analyses of ultrabasic rocks experiencing different degrees of serpentinization.

The locations of points corresponding to the compositions of oceanic hyperbasites, converted to coordinates of the nomogram according to the data in Table 1 and previously published data [2] are shown in Figure 2. A total of 71 analyses was used here, 54 of which characterize the Indian Ocean Ridge (circles) and 17 the Mid-Atlantic Ridge (crosses). It is easy to see that the hyperbasites of both oceans occupy practically the same narrow zone, lying in the field between the extreme theoretical lines of isochemical serpentinization and parallel to them. The very nature of the distribution of the points in Figure 2 indicates that serpentinization of oceanic hyperbasites

proceeded without notable addition or removal of bases or silicic acid and that the limits of change of the compositions of these rocks during serpentinization were practically the same for both ridges.

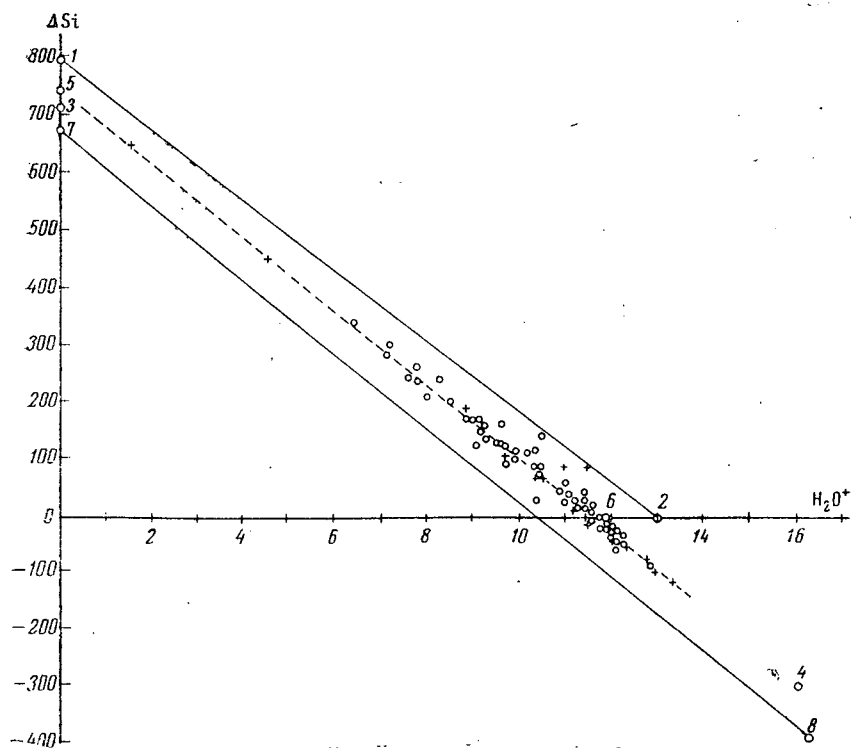


Figure 2. Graph of Change of ΔSi During Serpentinization of Oceanic Hyperbasites. Circles, Indian Ocean hyperbasites; Crosses, Atlantic Ocean hyperbasites. The broken line shows the position of the line corresponding to equations found by the method of least squares for the given series of analyses.

For mathematical proof of the isochemistry of serpentinization of oceanic hyperbasites, it is essential to show that the distribution of the analytic results in the coordinates of the nomogram is described by a linear equation and that the angular coefficient of this equation is sufficiently close to the angular coefficients of the theoretical lines of isochemical serpentinization.

The equation of a straight line approximating the results of the analyses was derived by the method of least squares³. This line is depicted in Figure 2 by the broken line. The equation has the form

$$\Delta\text{Si} + 63.75\text{H}_2\text{O}^+ = 747.4 = 0. \quad (7)$$

The coefficient of correlation between ΔSi and H_2O^+ was calculated for the purpose of assessing the quality of approximation. The calculated value is $R = -0.989$ (for a number of points $N = 71$).

It is known that the entire variability of the dependent variable (ΔSi) breaks down into variability which is random and that which is governed by a linear dependence (of ΔSi on H_2O^+). A measure of the latter when using the method of least squares is the coefficient of correlation. Here the share of the linear components in the variability of ΔSi is $R^2 \cdot 100 = 97.8\%$. This means that the dependence of ΔSi on H_2O^+ is practically functional (linear). The remaining share of the variability of ΔSi amounts to only 2.2%.

The error in the calculation of the coefficient of angular slope ΔK and free term Δa does not exceed $1 - R^2 / \sqrt{n} = 0.4\%$. Comparison of these results with the accuracy of individual determinations ($\Delta\text{Si} \approx 1\%$, $\text{H}_2\text{O}^+ \approx 5\%$) indicates that the probable variability of the path of the curve according to equation (7) falls within the limits of error of the analysis.

The equation of the top theoretical line of isochemical serpentinization (line 1-2) has the form:

$$\Delta\text{Si} + 61.23\text{H}_2\text{O}^+ = 796 = 0, \quad (8)$$

and that of the bottom theoretical line (line 7-8):

$$\Delta\text{Si} + 65.03\text{H}_2\text{O}^+ - 676 = 0. \quad (9)$$

³The calculations were done on the "Mir-1" computer, Department of Geochemistry, Moscow State University.

The average angular coefficient for equations (8) and (9) is 63.13, and for equation (7), 63.75. The relative difference between them does not exceed 1%, which shows it to be of random character.

Thus, it can be considered mathematically proven that serpentinization of the investigated oceanic hyperbasites proceeded isochemically -- with the addition of water along -- or that the processes of allometamorphic serpentinization were not notably manifested in these rocks.

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